Characteristics of propagation and scattering of light in nematic liquid crystals

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A study is made of the scattering of light and extinction in nematic liquid crystals allowing for the optical anisotropy. An expression is obtained for the intensity of the scattered light and the geometries in which there is no scattering by fluctuations of the director are considered. An analysis is made of the case of a weak optical anisotropy. It is shown that in some cases a relatively weak anisotropy may result in a considerable redistribution of the intensity of the scattered light. Calculations are made of the extinction coefficient and it is shown that the coefficient of an extraordinary ray is several times higher than that of an ordinary ray, and it depends on the angle between the ray and the director.

Nematic liquid crystals are uniaxial in respect of their optical properties. They differ from conventional (solid uniaxial crystals by strong fluctuations of the director orientation. The Fourier components of these fluctuations are of the same form as at a second-order phase transition point, which gives rise to a logarithmic divergence of the total scattering cross section at low angles. The extinction coefficient calculated in the first approximation for the kernel of a polarization operator depends on the dimensions of a sample.[1]

Investigations of the scattering of light in the nematic phase are usually carried out in the approximation of an optically isotropic medium.[2] However, the optical anisotropy of the majority of nematic liquid crystals is not weak[3] and it gives rise to a number of specific amounts. For example, an investigation of the extinction coefficient of nematic liquid crystals showed[4] that after allowance for the optical anisotropy the divergence of the total scattering cross section disappears for certain geometries. The extinction coefficient found experimentally for MBBA liquid crystals in three such geometries was found to be 5–15 cm^{-1}, depending on the geometry. Moreover, the conditions of propagation of the ordinary and extaordinary rays were quite different. The ordinary ray passed along all directions without distortion, whereas the extraordinary ray was converted from coherent to diffuse as a result of a strong forward scattering and this was accompanied by a considerable broadening of the ray. The effect depended on the thickness of the sample and on the angle between the ray and the director.[5]

We shall carry out a general analysis of the propagation and scattering of light in a nematic phase. We shall show that, in particular, even a relatively weak optical anisotropy of the medium can cause considerable changes in the scattering intensity for specific experimental geometries.

In the case of the ordinary ray we shall show that the total scattering cross section is finite for any direction of propagation and we shall calculate the extinction coefficient. We shall also show that in the case of the extraordinary ray the total cross section is finite only for the propagation along or across the director. For all other directions the total scattering cross section still diverges. The extinction coefficient can then be calculated in a single-loop approximation for the kernel of a polarization operator allowing for the finite size of the sample. It exhibits a strong angular dependence and is approximately an order of magnitude higher than the extinction coefficient of the ordinary ray.

1. GREEN FUNCTION OF AN ELECTROMAGNETIC FIELD IN AN ANISOTROPIC MEDIUM

Since strong fluctuations of the orientation of the director should result in significant scattering losses, a correct description of the propagation and scattering of light in nematic liquid crystals should allow not only for the optical anisotropy of the scattering medium, but also for the influence of fluctuations of the permittivity on the propagation of light.

It is known[6] that a consistent allowance for the correlation effects in the problem of propagation of light makes the effective permittivity ε of the medium a nonlocal quantity:

$$\varepsilon_{\alpha\beta}(\omega, \mathbf{k})=\varepsilon_{\alpha\beta}'(\omega, \mathbf{k})+i\varepsilon_{\alpha\beta}''(\omega, \mathbf{k}), \tag{1}$$

where $\mathbf{k}$ is the wave vector; ω is the angular frequency; $\hat{\varepsilon}'$ and $\hat{\varepsilon}''$ are real tensors. If we assume that the intrinsic absorption by the medium is weak, then the term ε'' in Eq. (1) describes the wave attenuation which is entirely due to the light losses as a result of the scattering, i.e., it is governed by the fluctuation contribution to $\hat{\varepsilon}$. In the case of non gyrotropic media, which include nematic liquid crystals, the tensors $\hat{\varepsilon}'$ and $\hat{\varepsilon}''$ are by definition symmetric.

In the case of a homogeneous medium it follows from the system of the Maxwell equations that

$$\left[k^2(\delta_{\alpha\beta}-s_\alpha s_\beta)-\left(\frac{\omega}{c}\right)^2\varepsilon_{\alpha\beta}\right]E_\beta=4\pi\left(\frac{\omega}{c}\right)^2 P_\alpha, \tag{2}$$

where $s_\alpha s_\beta = k_\alpha k_\beta/k^2$; c is the velocity of light in vacuum; $\mathbf{E}$ is the electric field intensity; $\mathbf{P}$ is the polarization vector. We shall define the Green function $T_{\alpha\beta}$ of an electromagnetic field in the medium by

$$E_\alpha=4\pi T_{\alpha\beta}P_\beta. \tag{3}$$

Here, the factor 4π is introduced for convenience in further calculations. We can find $\hat{T}$ simply by inverting the matrix on the left-hand side of Eq. (2). If the condition $\det\varepsilon\neq 0$ is satisfied, then $T_{\alpha\beta}$ can be represented in the form

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$$T_{\alpha\beta}=\sum_{j=1}^{2}\frac{k^2_{(j)}}{k^2-k^2_{(j)}}\frac{E^{(j)}_\alpha E^{(j)}_\beta}{(\mathbf{E}^{(j)}\hat{\varepsilon}\mathbf{E}^{(j)})},\tag{4}$$

where

$$k^2_{(i)}=\left(\frac{\omega}{c}\right)^2 n^2_{(i)}=\left(\frac{\omega}{c}\right)^2\frac{(\mathbf{E}^{(i)}\hat{\varepsilon}^2\mathbf{E}^{(i)})}{(\mathbf{E}^{(i)}\hat{\varepsilon}\mathbf{E}^{(i)})},$$
$$\mathbf{E}^{(i)}=\hat{\varepsilon}^{-1}\mathbf{D}^{(i)},\quad i=1,2,$$
$$k_{(3)}=\infty,\quad \mathbf{E}^{(3)}/E^{(3)}=\mathbf{s}.\tag{5}$$

Here $\mathbf{D}^{(i)}$ are the eigenvectors of a two-dimensional symmetric tensor of second rank $\varepsilon^{-1}_{\alpha\beta}$ considered in a plane orthogonal to the vector $\mathbf{s}$:

$$(\delta_{\alpha\beta}-s_\alpha s_\beta)\varepsilon_{\beta\gamma}^{-1}D_\gamma^{(i)}=\mu_{(i)}D_\alpha^{(i)},\tag{6}$$

where $\mu_{(i)}$ are the corresponding eigenvalues; $i=1, 2$. A complete convolution of the vectors $\mathbf{f}$, $\mathbf{g}$, with the tensor $\hat{t}$ will be described by $f_\alpha t_{\alpha\beta}g_\beta=(\mathbf{f}\hat{t}\mathbf{g})$. Lax and Nelson[7] obtained Eq. (4) for the case when $\varepsilon_{\alpha\beta}=\varepsilon_{\alpha\beta}(\omega)$; this formula also follows from the results of Ginzburg[8] on the emission of radiation by a charge in an anisotropic medium.

It should be noted that if $\mathbf{s}$ is a real vector, then in the uniaxial case when there is a preferred vector $\mathbf{n}$, the vectors $\mathbf{D}^{(i)}$ in Eq. (6) are real and one of them is coplanar with the vectors s and n, whereas the other is orthogonal to these vectors.[9]

The attenuation coefficient for the intensity of a plane wave with a wave vector $\mathbf{k}=\mathbf{k}'+i\mathbf{k}''$ is

$$\tau(\mathbf{k})=2k'',\tag{7}$$

where $\mathbf{k}$ is a root of the dispersion equation

$$(\mathbf{s}\hat{\varepsilon}\mathbf{s})(k^2-k^2_{(1)})(k^2-k^2_{(2)})=0.\tag{8}$$

The factor $(\mathbf{s}\hat{\varepsilon}\mathbf{s})$ in Eq. (8) corresponds to longitudinal waves, which we henceforward will ignore.

If we assume that the inequality $\hat{\varepsilon}''\ll\hat{\varepsilon}'$ is satisfied in Eq. (1), then in the first order with respect to $\hat{\varepsilon}''$ we can describe the quantity $k^2_{(i)}$ in Eq. (5) by[1)]

$$k^2_{(i)}=(\omega/c)^2(\mathbf{e}_0^{(i)}\hat{\varepsilon}\mathbf{e}_0^{(i)})/\cos^2\delta_0^{(i)},\tag{9}$$

where

$$\cos^2\delta^{(i)}=\frac{(\mathbf{e}^{(i)}\hat{\varepsilon}\mathbf{e}^{(i)})^2}{(\mathbf{e}^{(i)}\hat{\varepsilon}^2\mathbf{e}^{(i)})},\quad \mathbf{e}^{(i)}=\frac{\mathbf{E}^{(i)}}{E^{(i)}}.\tag{10}$$

The index "0" (used as a subscript or superscript) means that the corresponding quantity in Eqs. (5) and (10) is calculated for $\varepsilon''=0$. Then, $\delta_0^{(i)}$ is the angle between $\mathbf{E}_0^{(i)}$ and $\mathbf{D}_0^{(i)}=\hat{\varepsilon}_0\mathbf{E}_0^{(i)}$. Equations (7) and (8) yield the following expression for the attenuation coefficients of normal waves in the investigated medium:

$$\tau_{(i)}(\omega,\mathbf{k})=\left(\frac{\omega}{c}\right)^2\frac{(\mathbf{e}_0^{(i)}\hat{\varepsilon}''(\omega,\mathbf{k})\mathbf{e}_0^{(i)})}{k_{0(i)}\cos\chi\cos^2\delta_0^{(i)}}.\tag{11}$$

Here, χ is the angle between $\mathbf{k}'$ and $\mathbf{k}''$. In the case of homogeneous waves, which we shall henceforth consider, we have $k'\|k''$ *and* $\cos\chi=1$.

In the case of a uniaxial medium we find that neglect of the fluctuation contribution gives the following expression for the real part of $\hat{\varepsilon}$[1,2]

$$\varepsilon_{\alpha\beta}=\varepsilon_{0,\alpha\beta}=\varepsilon_\perp\delta_{\alpha\beta}+\varepsilon_a n_\alpha n_\beta.\tag{12}$$

In this case, we have

$$\mathbf{e}_0^{(1)}=\frac{[\mathbf{sn}]}{\sin\psi},\quad \mathbf{e}_0^{(2)}=\frac{\mathbf{s}\varepsilon_\|\cos\psi-\mathbf{n}(\varepsilon_\|\cos^2\psi+\varepsilon_\perp\sin^2\psi)}{\sin\psi(\varepsilon_\|^2\cos^2\psi+\varepsilon_\perp^2\sin^2\psi)^{1/2}},$$
$$\cos\delta_0^{(1)}=1,\quad \cos\delta_0^{(2)}=\frac{\varepsilon_\|\cos^2\psi+\varepsilon_\perp\sin^2\psi}{(\varepsilon_\|^2\cos^2\psi+\varepsilon_\perp^2\sin^2\psi)^{1/2}},\tag{13}$$
$$\cos\psi=\mathbf{sn}.$$

Here, $\varepsilon_a=\varepsilon_\|-\varepsilon_\perp$, where $\varepsilon_\|$ and $\varepsilon_\perp$ are the permittivities along and across $\mathbf{n}$. It is known that the solutions of the dispersion equation (8) are then $\mathbf{k}_{(1)}$ and $\mathbf{k}_{(2)}$ representing the ordinary and extraordinary waves, and we shall use the indices (1) and (2) (superscripts or subscripts) to identify these waves.

The problem of light scattering can be solved if we have an explicit expression for $T_{\alpha\beta}(\omega,\mathbf{R})$ at large distances $\mathbf{R}$. We can calculate $\hat{T}$ conveniently in the coordinate representation using the stationary phase method. A consistent application of this method to the Green fuction of an anisotropic medium, allowing for the curvature of the wave-vector surface at the stationary phase point was made in Ref. 10. It was found that in the case of a uniaxial medium the required expression is

$$T_{\alpha\beta}(\omega,\mathbf{R})=\frac{1}{4\pi R}\left(\frac{\omega}{c}\right)^2\sum_{i=1}^{2}n^2_{0(i)}\frac{e_{0\alpha}^{(i)}e_{0\beta}^{(i)}f_{0(i)}}{(\mathbf{e}_0^{(i)}\hat{\varepsilon}_0\mathbf{e}_0^{(i)})}\exp(i\mathbf{k}_{st}^{(i)}\mathbf{R}),\tag{14}$$

where

$$\mathbf{k}_{st}^{(1)}=\varepsilon_\perp^{1/2}\frac{\omega}{c}\frac{\mathbf{R}}{R},\quad \mathbf{k}_{st}^{(2)}=\frac{\omega}{c}\left[\frac{\varepsilon_\|\varepsilon_\perp}{(\mathbf{R}\hat{\varepsilon}_0^{-1}\mathbf{R})}\right]^{1/2}\hat{\varepsilon}_0^{-1}\mathbf{R}\tag{15}$$

are the vectors of the steady-state phase of the ordinary and extraordinary waves, and

$$n^2_{0(1)}=\varepsilon_\perp,\quad n^2_{0(2)}=\varepsilon_\|\varepsilon_\perp/(\mathbf{s}\hat{\varepsilon}_0\mathbf{s}),$$
$$f_{0(1)}=1,\quad f^2_{0(2)}=(\mathbf{s}\hat{\varepsilon}_0\mathbf{s})(\mathbf{s}\hat{\varepsilon}_0^2\mathbf{s})/\varepsilon_\|\varepsilon_\perp^2.\tag{16}$$

All the quantities $n_{0(i)}$, $e_0^{(i)}$, $f_{0(i)}$ in Eq. (16) are calculated for the appropriate vector of the steady-state phase $\mathbf{k}=\mathbf{k}_{si}^{(i)}$, where $i=1, 2$.

2. SCATTERING OF LIGHT IN A NEMATIC LIQUID CRYSTAL WITH AN OPTICAL ANISOTROPY

The scattering of light occurs on fluctuations of the permittivity tensor $\delta\varepsilon_{\alpha\beta}$. Using Eq. (14), we can readily obtain the expression for the intensity of light scattered in a uniaxial anisotropic meidum[10]:

$$I_\beta^\alpha(\mathbf{R})=I^{(i)}\frac{V}{R^2}\left(\frac{\omega^2}{4\pi c^2}\right)^2\frac{1}{n_{0(i)}\cos\delta_0^{(i)}}\sum_{j=1}^{2}\frac{n_{0(j)}}{\cos^3\delta_0^{(j)}}$$
$$\times f^2_{0(j)}(e_{0\beta}^{(j)})^2 e_{0\nu}^{(i)}e_{0\mu}^{(i)}\langle\delta\varepsilon_{\nu\alpha}\delta\varepsilon_{\mu\alpha}^*\rangle_{\mathbf{q}(i,j)},\tag{17}$$

where $I^{(i)}$ is the intensity (modulus of the Poynting vector) of the incident light; V is the scattering volume; $\mathbf{R}/R$ is the direction toward an observation point separated from the scattering volume by a distance $R \gg V^{1/3}$; α and β are the polarizations of the incident and scattered light which are not summed in Eq. (17); $\mathbf{q}(i, j) = \mathbf{k}_{s(j)} - \mathbf{k}_{i(i)}$, where $\mathbf{k}_{s(j)} = \mathbf{k}(\mathbf{R}/R)$ is the wave vector of the scattered light and $\mathbf{k}_{i(i)}$ is the wave vector of the incident plane wave; the angular brackets $\langle ... \rangle$ denotes statistical averaging. The quantities with the index (j) are calculated for the wave vector $k_{s(j)}$ and those with the index (i) are calculated for the wave vector $k_{i(i)}$. The formula (17) is derived on the assumption that the incident and scattered waves travel inside the medium. The effects associated with the refraction and the boundaries of a sample were discussed in detail in Ref. 10. Equation (17) was derived using the following expression for the modulus of the Poynting vector in an anisotropic medium:

$$I^{(i)} = (c/4\pi)\,|\mathbf{E}_0^{(i)}|^2 n_{0(i)} \cos\delta_0^{(i)},$$

where $\mathbf{E}_0^{(i)}$ is the intensity of the electric field; use was also made of the relationship $n_{(i)}^2 \cos^2\delta^{(i)} = (\mathbf{e}^{(i)}\hat{\varepsilon}\mathbf{e}^{(i)})$, and of the fact that in the case of a spatially inhomogeneous medium, we have

$$\langle \delta\varepsilon_{\alpha\beta,\,\mathbf{q}_1} \delta\varepsilon_{\gamma\delta,\,\mathbf{q}_2} \rangle = \delta(\mathbf{q}_1 + \mathbf{q}_2) \langle \delta\varepsilon_{\alpha\beta} \delta\varepsilon_{\gamma\delta}^* \rangle_{\mathbf{q}_1}. \tag{18}$$

In the nematic phase the correlation function $G_{\alpha\beta\gamma\delta}(\mathbf{q}) = \langle \delta\varepsilon_{\alpha\beta}\delta\varepsilon^*_{\gamma\delta} \rangle_{\mathbf{q}}$ includes contributions from Eq. (17) and these contributions represent fluctuations of three types: fluctuations of the director ξ_1 and ξ_2, biaxial fluctuations ξ_3 and ξ_4, and longitudinal fluctuations σ (Refs. 11–13). Since the strongest fluctuations are those of the director, they can be discovered against the background of weaker ξ_3, ξ_4, and σ modes if the experimental geometry is such that it suppresses the contributions of ξ_1 and ξ_2 to the scattering. The problem was considered in Refs. 11–13 for the case when $\varepsilon_a = 0$ and the conditions for the absence of scattering by ξ_1 and ξ_2 were found to be

$$\alpha\mathbf{n} = \beta\mathbf{n} = 0, \quad (\alpha \pm \beta) \parallel \mathbf{n}. \tag{19}$$

We can easily show that in the case of an anisotropic medium the relevant conditions are given by Eq. (19). The only difference is that α and β should coincide with the allowed directions of the polarizations in the anisotropic medium. As a result of this restriction, the conditions of Eq. (19) are obeyed in the following cases:

$$1)\ \alpha\mathbf{n} = \beta\mathbf{n} = 0;\quad 2)\ \alpha \parallel \beta \parallel \mathbf{n},\ \mathbf{k}_i\mathbf{n} = \mathbf{k}_s\mathbf{n} = 0;$$

$$3)\ \mathbf{n} \parallel \mathbf{k}_i \pm \mathbf{k}_s,\ \alpha, \beta \in Q_0, \tag{20}$$

where Q_0 is a plane containing $\mathbf{k}_i$ and $\mathbf{k}_s$. It is interesting to note that the condition 1) of Eq. (20) in fact means that if the incident beam is ordinary, then the whole light scattered by the director fluctuations has the extraordinary polarization.

Using the expression for the correlation function $G_{\alpha\beta\gamma\delta}(\mathbf{q})$ found in Ref. 13 and also Eq. (17), we readily obtain the following expressions for the scattering intensities in the case of these three geometries:

$$1)\ I_\beta{}^\alpha = \frac{I_0 V}{R^2}\left(\frac{\omega^2}{4\pi c^2}\right)^2 \Big\{ \langle |\xi_3|^2 \rangle \sin^{-2}\varphi_0 [p_\alpha{}^2 + p_\beta{}^2 + 2\alpha\beta p_\alpha p_\beta - 4p_\alpha{}^2 p_\beta{}^2 \sin^{-2}\varphi_0]$$

$$+ \left\langle \left| \xi_4 [2p_\alpha p_\beta \sin^{-2}\varphi_0 - \alpha\beta] - \frac{1}{3}\sigma\alpha\beta \right|^2 \right\rangle \Big\};$$

$$2)\ I_\beta{}^\alpha = \frac{I_0 V}{R^2}\left(\frac{\omega^2}{4\pi c^2}\right)^2 \frac{\varepsilon_\perp}{\varepsilon_\parallel} \cdot \frac{4}{9} \langle |\sigma|^2 \rangle;$$

$$3)\ I_\beta{}^\alpha = \frac{I_0 V}{R^2}\left(\frac{\omega^2}{4\pi c^2}\right)^2 \frac{\varepsilon_\parallel{}^2 + \varepsilon_\perp{}^2 \pm (\varepsilon_\parallel{}^2 - \varepsilon_\perp{}^2)\cos\theta_0}{\varepsilon_\parallel \varepsilon_\perp{}^2 (\varepsilon_\parallel + \varepsilon_\perp \pm \varepsilon_a \cos\theta_0)^3}$$

$$\times \left\langle \Big| \xi_4 \varepsilon_\parallel{}^2 (1 \pm \cos\theta_0) \mp \frac{1}{3}\sigma [2\varepsilon_\perp{}^2 + \varepsilon_\parallel{}^2 \pm (\varepsilon_\parallel{}^2 - 2\varepsilon_\perp{}^2)\cos\theta_0] \Big|^2 \right\rangle, \tag{21}$$

where I_0 is the intensity of the incident light; $\mathbf{p} = \mathbf{q}/q$; $\cos\varphi_0 = \mathbf{p}\cdot\mathbf{n}$; θ_0 is the scattering angle formed by the vectors $\mathbf{k}_i$ and $\mathbf{k}_s$.

The degree to which the geometric conditions of Eq. (20) are obeyed is determined by the inequality

$$R_{sc}(\xi_1, \xi_2) \left[\frac{\delta}{2\sin(\theta_0/2)}\right]^2 < \frac{1}{6} R_{sc}(\xi_3, \xi_4, \sigma)\,(\alpha\beta)^2,$$

where

$$R_{sc}(\xi_1, \xi_2) \approx \left(\frac{\omega^2}{4\pi c^2}\right)^2 \frac{k_B T \varepsilon_a^2}{K_{ii} k^2}$$

is the constant for the scattering by the director fluctuations; k_b is the Boltzmann constant; K_{ll} is a Frank modulus; δ is the angle of deviation of α, β, and $\mathbf{n}$ from the vectors which satisfy exactly the conditions of Eq. (20); $R_{sc}(\xi_3, \xi_4, \sigma)$ is the scattering constant related to fluctuations of the quantities ξ_3, ξ_4, and σ. If $\varepsilon_a \sim 1$ and $K_{ll} \sim 10^{-6}$ dyn, then $R_{sc}(\xi_1, \xi_2) \sim 1\ \text{cm}^{-1}$. Assuming that $R_{sc}(\xi_3, \xi_4, \sigma) \approx R_{sc}(\sigma) \sim 10^{-2}\ \text{cm}^{-1}$ (Ref. 14), we obtain $\delta \leqslant 1.5°$.

Even when the conditions of Eq. (20) are satisfied exactly, fluctuations of the director make a contribution to the scattering because of repeated absorption and reemission. The condition of smallness of this contribution is the inequality

$$R_{sc}{}^2(\xi_1, \xi_2)\,V^{1/3} < R_{sc}(\xi_3, \xi_4, \sigma)\,(\alpha\beta)^2,$$

which for $\varepsilon_a \sim 1$ limits the scattering volume to $V^{1/2} < 10^{-2}$ cm. This estimate is obtained for the geometry 1) in one-constant approximation for the Frank moduli allowing for the contribution of double scattering of light.[15] The influence of extinction is ignored: $V^{1/2}\tau < 1$.

It follows from the above estimates that the contributions described by the expressions in Eq. (21) are easiest to detect by selecting nematic liquid crystals with the smallest values of ε_a.

3. CASE OF A WEAK OPTICAL ANISOTROPY

If the optical anisotropy of a nematic liquid crystal is weak, so that $|\varepsilon_a| \ll \varepsilon_\parallel, \varepsilon_\perp$, it is usual to assume that the intensity of the scattered light is always the same as in the case of an isotropic medium.[2] Since we then have $\cos\delta^{(2)} = 1$ and

$n_{(1)} = n_{(2)}$, it follows that in the case of a purely ordinary or a purely extraordinary ray Eq. (17) does indeed reduce to the formula for the scattering in an isotropic medium. However, if we consider the geometry when both rays travel in a medium, then the intensities of the scattering in an isotropic medium and in a medium with a weak optical anisotropy may differ quite considerably.

If the divergence between the ordinary and extraordinary rays formed as a result of the incident of light on the boundary of a crystal can be ignored compared with the ray diameters, then the electric field vector in an anisotropic medium is given by

$$\mathbf{E}(\mathbf{r}) = \alpha^{(1)}E^{(1)}\exp\{i\mathbf{k}_{i(1)}\mathbf{r}\} + \alpha^{(2)}\mathbf{E}^{(2)}\exp\{i\mathbf{k}_{i(2)}\mathbf{r}\}, \quad (22)$$

where $\boldsymbol{\alpha}^{(1)}$ and $\boldsymbol{\alpha}^{(2)}$ are the polarization vectors of the ordinary and extraordinary waves; $E^{(1)}$ and $E^{(2)}$ are the amplitudes of these waves; $\mathbf{k}_{i(1)}/k_{i(1)} = \mathbf{k}_{i(2)}/k_{i(2)}$ is the direction of propagation of the waves.

The field of a wave scattered in a medium with a weak anisotropy can be represented by the following expression which is derived from Eqs. (3) and (14):

$$E_\beta(\mathbf{R}) = \frac{1}{4\pi R}\left(\frac{\omega}{c}\right)^2 \sum_{l,j=1}^{2} E^{(l)}e_\beta^{(j)}e_\mu^{(j)}\alpha_\nu^{(l)}\,\delta\varepsilon_{\mu\nu,\mathbf{k}_{s(j)}-\mathbf{k}_{i(l)}} \times \exp\{ik_{s(j)}R\} \quad (23)$$

In the case of anisotropic medium, we have

$$\mathbf{k}_{i(1)} = \mathbf{k}_{i(2)} = \mathbf{k}_i, \qquad \mathbf{k}_{s(1)} = \mathbf{k}_{s(2)} = \mathbf{k}_s, \quad (24)$$

and $\mathbf{E}_\beta(\mathbf{R})$ obtained using the relationship

$$e_\nu^{(1)}e_\mu^{(1)} + e_\nu^{(2)}e_\mu^{(2)} + k_{p,\nu}k_{p,\mu}/k^2 = \delta_{\nu\mu}$$

assumes its usual form

$$E_\beta(\mathbf{R}) = \frac{E}{4\pi R}\left(\frac{\omega}{c}\right)^2 \times\left(\delta_{\beta\mu} - \frac{k_{p,\beta}k_{p,\mu}}{k^2}\right)\delta\varepsilon_{\mu\nu,\mathbf{k}_s-\mathbf{k}_i}\alpha_\nu\, exp(ik_sR), \quad (25)$$

where

$$E = [E^{(1)2} + E^{(2)2}]^{1/2}, \quad \alpha = [\alpha^{(1)}E^{(1)} + \alpha^{(2)}E^{(2)}]/E. \quad (26)$$

In an isotropic medium the vectors $\mathbf{e}^{(1)}$ and $\mathbf{e}^{(2)}$ can be regarded as arbitrary and it is assumed that they form a set of three unit vectors with $\mathbf{k}_s/k_s$.

We must draw attention to the fact that in calculation of the intensity of the scattered light in an anisotropic medium, it follows from Eq. (18) that in the quantities $\langle E_\beta(\mathbf{R})\mathbf{E}_\beta^*(\mathbf{R})\rangle$ which are being averaged only the terms $\langle\delta\varepsilon_{q1}, \delta\varepsilon_{-q2}\rangle$ differ from zero and these have identical vectors $\mathbf{q} = \mathbf{k}_{s(j)} - \mathbf{k}_{i(l)}$. Then, in the case of a medium with a weak but finite anisotropy we generally have, when neither $\mathbf{k}_i$ nor $\mathbf{k}_s$ are directed along $\mathbf{n}$, we have

$$\langle E_\beta(\mathbf{R})E_\beta^*(\mathbf{R})\rangle = \left(\frac{\omega^2}{4\pi Rc^2}\right)^2 \sum_{l,j=1}^{2} |E^{(l)}|^2 (e_\beta^{(j)})^2 \times e_\mu^{(j)}e_\nu^{(j)}\langle\delta\varepsilon_{\mu\varphi,\mathbf{k}_{s(j)}-\mathbf{k}_{i(l)}}\delta\varepsilon_{\nu\psi,\mathbf{k}_{i(l)}-\mathbf{k}_{s(j)}}\rangle\alpha_\varphi^{(l)}\alpha_\psi^{(l)}. \quad (27)$$

In the case of an isotropic medium, we find from Eqs. (25) and (26) that

$$\langle E_\beta(\mathbf{R})E_\beta^*(\mathbf{R})\rangle = \left(\frac{\omega^2}{4\pi Rc^2}\right)^2 \sum_{j_1,l_1=1}^{2}\sum_{j_2,l_2=1}^{2} E^{(l_1)}E^{(l_2)*}e_\beta^{(j_1)}e_\beta^{(j_2)}e_\mu^{(j_1)}e_\nu^{(j_2)} \times\langle\delta\varepsilon_{\mu\varphi,\mathbf{k}_{s(j_1)}-\mathbf{k}_{i(l_1)}}\delta\varepsilon_{\nu\psi,\mathbf{k}_{i(l_2)}-\mathbf{k}_{s(j_2)}}\rangle\alpha_\varphi^{(l_1)}\alpha_\psi^{(l_2)}. \quad (28)$$

It follows from Eq. (24) that all the correlation functions in Eq. (28) are finite and Eq. (27) contains only four out of six in terms that occur in Eq. (28) and these terms are characterized by $l_1 = l_2$ and $j_1 = j_2$. This naturally can give rise to large differences beween the scattering intensities in an isotropic medium and in a medium with a weak anisotropy. The difference between Eqs. (17) and (28) can be of either sign. If $\boldsymbol{\beta} = \mathbf{e}^{(1)}$ or $\mathbf{e}^{(2)}$ and $\boldsymbol{\alpha} = \boldsymbol{\alpha}^{(1)}$ or $\boldsymbol{\alpha}^{(2)}$, then Eqs. (27) and (28) are identical.

It should be pointed out that if $\mathbf{k}_i \| \mathbf{n}$, then $\mathbf{k}_{i(1)} = \mathbf{k}_{i(2)}$, and it follows from Eqs. (24) and (18) that Eq. (27) acquires four additional terms and the complete expression contains eight terms from Eq. (28), where $j_1 = j_2$. A similar situation occurs also if $\mathbf{k}_s \| \mathbf{n}$. In this case eight terms of Eq. (28) with $l_1 = l_2$ are retained. Finally, if $\mathbf{k}_i \| \mathbf{k}_s \| \mathbf{n}$, the contribution to the scattering made for $\varepsilon_a \neq 0$ comes from all sixteen terms of Eq. (28). Consequently, the scattering indicatrix of an anisotropic medium may have peaks and dips along the selected directions of $\mathbf{k}_i$ and $\mathbf{k}_s$, i.e., even a weak anisotropy may give rise to a finite redistribution of the light-scattering intensity.

This conclusion is based on Eq. (18), which is rigorously valid only for an infinite volume. When the volume is finite and given by $V \propto L^3$, the δ function in Eq. (18) can be replaced by its approximation with a finite width of the order of $1/L$. Therefore, the weakest optical anisotropy for which Eq. (27) becomes valid is limited by the inequality $|\mathbf{k}_{(1)} - \mathbf{k}_{(2)}| \gg 1/L$ or $\Delta n = |n_{(1)} - n_{(2)}| \gg \lambda/L$, where λ is the wavelength of light. If the thickness of a liquid crystal is $L \sim 0.1$ cm, then in the visible part of the spectrum this leads to the restriction $\Delta n \gg 10^{-3}$, which is known to be satisfied by nematic liquid crystals.[3] Therefore, neglect of the optical anisotropy in any real nematic liquid crystal may in some geometries give rise to an error which is of the same order as the intensity of the scattered light itself.

It should be stessed that Eq. (27) is derived on the assumption that the divergence beween the ordinary and extraordinary rays created by a single external beam incident on the boundary of the crystal is small, which imposes an upper limit on Δn: $\Delta n \ll rn/L$, where r is the ray diameter. Our discussion is valid if

$$\lambda/L \ll \Delta n \ll rn/L. \quad (29)$$

However, it should be pointed out that, for example, in the geometry in which the director is orthogonal to the scattering plane, there is no divergence beween the ordinary and extraordinary rays for light incident normally on the boundary of a crystal[1] and in this case there is no upper limit set by Eq. (29).

It should be noted that a redistribution of the intensity of the scattered light may, in principle, be also observed in the case of an isotropic medium when a weak anisotropy is created in this medium by, for example, the Kerr effect or the elastooptic effect.

For example, in the case of scattering by fluctuations of a scalar parameter with a correlation function $G_{\alpha\beta\gamma\delta} \propto \delta_{\alpha\beta}\delta_{\gamma\delta}$ in a geometry in which the optic axis is orthogonal to the scattering plane, the ratio of the scattering intensities in isotropic $I^{\alpha}_{\beta is}$ and anisotropic $I^{\alpha}_{\beta a}$ media is

$$I^{\alpha}_{\beta is}/I_{\beta a}{}^{\alpha}=(\alpha\beta)^2/[(\alpha\beta-n_\alpha n_\beta)^2+n_\alpha{}^2 n_\beta{}^2].$$

It follows in particular from this formula that if $\alpha\cdot\beta=0$, $n_\alpha\neq 0$, and $n_\beta\neq 0$, then $I^{\alpha}_{\beta a}\neq 0$ and $I^{\alpha}_{\beta is}=0$.

4. EXTINCTION COEFFICIENT

It follows from Eq. (12) that the extinction coefficient is governed by the imaginary part of the permittivity tensor $\varepsilon''_{\alpha\beta}(\omega,\mathbf{k})$. If we use the ladder approximation for the kernel of the intensity operator, we find that the nonlocal part of the permittivity is

$$\varepsilon_{\alpha\beta}(\omega,\mathbf{k})-\varepsilon_{\alpha\beta}{}^0=\frac{1}{(2\pi)^3}\int d\mathbf{q}\, T_{\nu\mu}(\mathbf{q})\, G_{\alpha\nu\beta\mu}(\mathbf{k}-\mathbf{q}). \quad (30)$$

The imaginary part of this relationship represents an optical theorem which relates the attenuation of an electromagnetic wave in a medium to the scattering properties of this medium.[6] If we ignore the longitudinal term[2)] in Eq. (4) and allow for Eq. (11), we find that for $\mathbf{k}=\mathbf{k}_{0(i)}$ this optical theorem becomes

$$\tau(\omega,\mathbf{k}_{0(i)})=\frac{1}{(2\pi)^3}\left(\frac{\omega}{c}\right)^4\frac{e^{(i)}_{0\alpha}e^{(i)}_{0\beta}}{k_{0(i)}\cos^2\delta^{(i)}_0}$$
$$\times\sum_{j=1}^{2}\int\frac{e^{(j)}_{0\nu}e^{(j)}_{0\mu}}{\cos^2\delta^{(j)}_0}\frac{k_{0(j)}\tau_{(j)}}{(q^2-k^2_{0(j)})^2+k^2_{0(j)}\tau^2_{(j)}}G_{\alpha\nu\beta\mu}(\mathbf{k}_{0(i)}-\mathbf{q})\,d\mathbf{q}. \quad (31)$$

Here all the quantities with the index (i) are functions of $\mathbf{k}_{0(i)}$ and the quantities with the index (j) are functions of $\mathbf{q}$. In the derivation of Eq. (31) we have replaced $\mathbf{e}^{(j)}$ and ε in the integrand by $\mathbf{e}_0^{(j)}$ and $\hat{\varepsilon}_0$, and we have used also the equality $k^2_{(j)}=k^2_{0(j)}+ik_{0(j)}\tau_{(j)}$, which is valid if $\tau_{(j)}/k_{0(j)}\ll 1$. The smallness of the latter ratio allows us in most cases to go to the limit $\tau_{(j)}\to 0$ on the right-hand side of Eq. (31), which effectively corresponds to the replacement of $\hat{T}$ by $\hat{T}_0$ in Eq. (30). If we use

$$\lim_{\varepsilon\to 0}\frac{\varepsilon}{x^2+\varepsilon^2}=\pi\delta(x),$$

then the resultant δ function reduces the three-dimensional integral to a surface integral, and we obtain the optical theorem in its usual form

$$\tau(\omega,k_{0(i)})=\left(\frac{\omega^2}{4\pi c^2}\right)^2\frac{e^{(i)}_{0\alpha}e^{(i)}_{0\beta}}{n_{0(i)}\cos^2\delta^{(i)}_0}\sum_{j=1}^{2}\int d\Omega^{(j)}_{\mathbf{q}}$$
$$\times\frac{n_{0(j)}e^{(j)}_{0\nu}e^{(j)}_{0\mu}}{\cos^2\delta^{(j)}_0}G_{\alpha\nu\beta\mu}(\mathbf{k}_{0(i)}-\mathbf{q}), \quad (32)$$

where $d\Omega^{(j)}_{\mathbf{q}}$ denotes integration over the surface $q=k_{0(j)}(\mathbf{q})$.

The main contribution to the scattering (and, consequently to the extinction) in a nematic liquid crystal comes from fluctuations of the director ξ_1 and ξ_2, which are characterized by the correlation function

$$G_{\alpha\beta\gamma\delta}(\mathbf{q})=\sum_{l=1}^{2}\langle|\xi_l|^2\rangle(e_{l\alpha}e_{l\gamma}n_\beta n_\delta+e_{l\alpha}e_{l\delta}n_\beta n_\gamma$$
$$+e_{l\beta}e_{l\delta}n_\alpha n_\gamma+e_{l\beta}e_{l\gamma}n_\alpha n_\delta), \quad (33)$$

where

$$\langle|\xi_l|^2\rangle=k_BT\varepsilon_a^2/[K_{ll}q^2+(K_{33}-K_{ll})(\mathbf{qn})^2],$$
$$\mathbf{e}_2=[\mathbf{qn}]/q\sin\theta, \quad (34)$$
$$\mathbf{e}_1=[\mathbf{e}_2\mathbf{n}],\quad \cos\theta=\mathbf{qn}/q.$$

It should be noted that the existence of a pole $1/q^2$ in Eq. (33) should result in a logarithmic divergence of the integrals in Eq. (32). However, if $\psi\neq 0$ between $\mathbf{k}_i$ and $\mathbf{n}$, the scattering vector $\mathbf{k}_{s(j)}-\mathbf{k}_{i(i)}$ corresponding to $j\neq i$ always differs from zero because $\mathbf{k}_{s(j)}\neq\mathbf{k}_{i(i)}$. It therefore follows that a divergent contribution to the integral (32) can come only from the scattered light with the same polarization as that of the incident light. However, this divergence may be absent if for geometric reasons the scattering through zero angle is finite when $i=j$. Therefore, the geometries in which the integrals over all the angles in Eq. (32) are finite can be identified from the requirement that one of the conditions for the absence of the scattering of light by the director fluctuations (20) is satisfied for the scattering by zero angle in the case of rays with the same polarizations for the incident and scattered light. It follows from Eq. (20) that this requirement is always satisfied if $\mathbf{k}_{i(i)}$ is the ordinary ray, whereas if $\mathbf{k}_{i(i)}$ is the extraordinary ray, this is true only if $\psi=\pi/2$. For $\psi=0$ the condition (20) is also satisfied, but this case applies to both the ordinary and extraordinary rays. We can easily see that all three geometries investigated in Ref. 4, i.e., $\mathbf{k}_i\|\mathbf{n}$, $\mathbf{k}_i\perp\mathbf{n}$ and $\alpha\perp\mathbf{n}$, $\mathbf{k}_i\perp\mathbf{n}$ and $\alpha\|\mathbf{n}$, belong to one of these types.

Substituting Eqs. (33) and (34) into Eq. (32) and integrating with respect to the azimuthal angle φ, we find that the extinction coefficient of the ordinary ray [in view of Eq. (20) a nonzero contribution to Eq. (32) is made only by the term with $j=2$] is described by the expression

$$\tau_{(1)\psi}=\frac{\omega^2}{8\pi c^2}\frac{k_BT}{K_{33}}\frac{\varepsilon_a^2}{(\varepsilon_\|\varepsilon_\perp)^{1/2}}\int_{-1}^{1}du\frac{1-u^2}{(1+au^2)^{1/2}}$$
$$\times\left[(1-u^2)\frac{t_2-t_1}{\Phi_1t_2+\Phi_2t_1}\left(\frac{t_1}{M+At_1+\Phi_1}+\frac{t_2}{M+At_2+\Phi_2}\right)+\frac{1}{\Phi_2}\right], \quad (35)$$

where

$$A=\rho^2\sin^2\psi+1-u^2,\quad M=(\rho\cos\psi-u)^2,$$
$$\Phi_l=[M^2+2t_lAM+t_l^2F^2]^{1/2},\quad F=\rho^2\sin^2\psi-1+u^2,$$
$$\rho=\left(\frac{1+au^2}{1+a}\right)^{1/2},\quad a=\frac{\varepsilon_a}{\varepsilon_\perp},\quad t_l=\frac{K_{ll}}{K_{33}},\quad l=1,2.$$

In the case of the extraordinary wave when $0 < \psi < \pi/2$, both terms in the sum (32) contribute to the extinction: $\tau_{(2)} = \tau_{(2)}^{(1)} + \tau_{(2)}^{(2)}$. The term with $j = 1$ is calculated by analogy with Eq. (35) and it is given by

$$\tau_{(2)}^{(1)}(\psi) = \frac{\omega^2}{8\pi c^2}\frac{k_B T}{K_{33}}\frac{\varepsilon_a^2}{(\varepsilon_\parallel\varepsilon_\perp)^{1/2}}\frac{\sin^2\psi}{(1+a\cos^2\psi)^{1/2}}$$
$$\times\int_{-1}^{1} du\left[\rho^2\sin^2\psi\frac{t_2-t_1}{\Phi_1 t_2+\Phi_2 t_1}\left(\frac{t_1}{M+At_1+\Phi_1}+\frac{t_2}{M+At_2+\Phi_2}\right)+\frac{1}{\Phi_2}\right], \quad (36)$$

where

$$\rho = [(1+a)(1+a\cos^2\psi)^{-1}]^{1/2}.$$

the same notation is used in the above formula as in Eq. (35), with the exception of ρ.

The term with $j = 2$ diverges in the limit of small scattering angles. If this contribution to $\tau_{(2)}$ is calculated, integration is carried out only for angles $\theta_{\min} \sim \lambda/L$, where L is a certain characteristic length. In particular, if a sample is completely illuminated, the $\theta_{\min}$ is the angle of diffraction and L is the size of the sample.[1] If a thin beam crosses the sample, we can easily show that L is the transverse size of the beam. Since the main contribution comes from the range of small angles, then in the approximation which is quadratic with respect to θ we obtain

$$\tau_2^{(2)}(\psi)$$
$$= \left(\frac{\omega}{4\pi c}\right)^2\frac{k_B T}{K_{33}}\frac{\varepsilon_a^2\varepsilon_\parallel\varepsilon_\perp}{P}\sin^2 2\psi\ln\frac{L}{\lambda}\int_0^{2\pi} d\varphi\frac{1}{P^2+Q\cos^2\varphi\sin^2\psi}$$
$$\times\left[\frac{\varepsilon_\parallel^2\cos^2\psi\cos^2\varphi}{t_1P^2+(t_1Q+\varepsilon_\perp^2)\cos^2\varphi\sin^2\psi}+\frac{P^2\sin^2\varphi}{t_2P^2+(t_2Q+\varepsilon_\perp^2)\cos^2\varphi\sin^2\psi}\right], \quad (37)$$

where

$$P = \varepsilon_\parallel\cos^2\psi+\varepsilon_\perp\sin^2\psi, \qquad Q = \varepsilon_a^2\cos^2\psi-\varepsilon_\perp^2.$$

Integrating with respect to φ, we obtain

$$\tau_{(2)}^{(2)} = \frac{\omega^2}{8\pi c^2}\frac{k_B T\varepsilon_a^2}{K_{33}}\frac{\varepsilon_\parallel\varepsilon_\perp\sin^2 2\psi}{(\varepsilon_\parallel\cos^2\psi+\varepsilon_\perp\sin^2\psi)^2}\frac{t_1(F_1+F_2)}{F_1(t_1F_2+t_2F_1)}$$
$$\times\ln\frac{L}{\lambda}, \quad (38)$$

where

$$F_i = (t_i^2\varepsilon_\parallel^2\cos^2\psi+t_i\varepsilon_\perp^2\sin^2\psi)^{1/2}, \quad i=1, 2.$$

the formulas for $\tau_{(1)}(0) = \tau_{(2)}(0)$, $\tau_{(1)}(\pi/2)$, and $\tau_{(2)}(\pi/2)$ can be found in Ref. 4.

In the case of typical nematic liquid crystals at $\varepsilon_a \sim 1$ and $K \sim 10^{-6}$ dyn when $L \sim 0.1$ cm, we find that

$$\tau_{(1)} \sim \frac{1}{8\pi}\left(\frac{\omega}{c}\right)^2\frac{k_B T}{K_{ll}}\frac{\varepsilon_a^2}{\varepsilon} \sim 5\ \text{cm}^{-1},$$
$$\tau_{(2)} \sim \tau_{(1)}\ln\frac{L}{\lambda} \sim 40\ \text{cm}^{-1}.$$

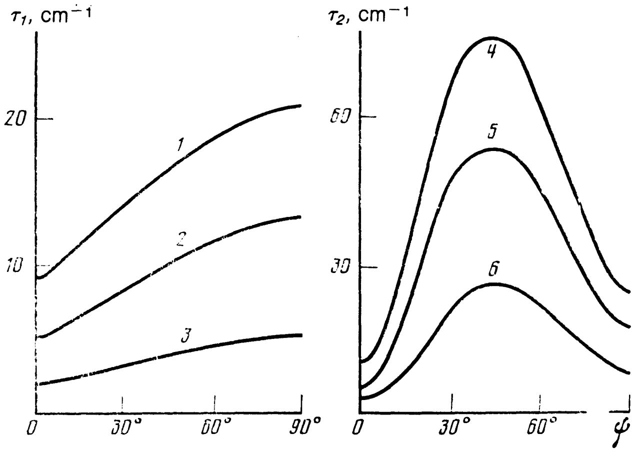

FIG. 1. Angular dependences of the extinction coefficients of the ordinary and extraordinary rays traveling in the following nematic liquid crystals: 1), 4) BMOAB (*p-n*-butyl-*p*-methoazoxybenzene); 2), 5) MBBA (4-methoxybenzylidene butylaniline); 3), 6) N-106.

Figure 1 shows the angular dependences of the extinction coefficients of the ordinary and extraordinary rays in three different liquid crystals. The values of K_{ll}, $\varepsilon_\parallel$, and $\varepsilon_\perp$ were taken from Refs. 3 and 16, whereas L was assumed[3)] to be 1 mm. The main contribution to $\tau_{(2)}$ comes from the term $\tau_2^{(2)}$. We can see that τ_2 depends strongly on the angle between the ray and director, and is several times greater than the extinction coefficient for the ordinary ray.

It should be pointed out that if the limit $\tau_{(j)} \to 0$ is not used on the right-hand side of Eq. (31), the integral over the angles becomes finite even for $j = 2$ and the cutoff parameter is the attenuation length $\tau_{(2)}^{-1}$. This method of avoiding the divergence was used in Ref. 17 to calculate the extinction coefficient at the critical point. However, if we bear in mind that, strictly speaking, $\tau = \tau(\omega, \mathbf{k})$, we have to carry out calculations for a complex value of $\mathbf{k} = \mathbf{k}_{(i)}$, which is a root of the dispersion equation, and not for $\mathbf{k} = \mathbf{k}_{0(i)}$, so that—as is readily established—the divergence in the angular integral for $\tau(\omega, \mathbf{k}_{(i)})$ is retained and the cutoff parameter is still $\theta_{\min}$.

In the presence of an external magnetic field H the correlation function acquires an energy gap $\chi_a H^2$: $G(q) \propto (q^2K_{ll} + \chi_a H^2)^{-1}$, where χ_a is the magnetic susceptibility.[2] The total scattering cross section then becomes finite, but a significant reduction in the extinction coefficient occurs when the "magnetic coherence length" becomes much less than the characteristic length L: $H^{-1}(K_{ll}/\chi_a)^{1/2} \ll L$. For typical values of $\chi_a \sim 10^{-7}$ and $K_{ll} \sim 10^{-6}$ dyn, and $L \sim 1$ mm, a reduction in the extinction coefficient by, for example, a factor of 2 occurs for $H \sim 10^3$ Oe. It should be stressed that such fields have practically no influence on the scattering intensity.

It should be pointed out that in view of the extreme elongation of the scattering indicatrix, almost all the light losses due to the scattering in the extraordinary ray occur because of the forward scattering. Consequently, the extraordinary ray is transformed because of multiple forward scattering from coherent to diffuse and it is broadened somewhat. This effect has been observed experimentally.[5]

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[1)]We shall show below that in the case of nematic liquid crystals we have $\varepsilon''/\varepsilon' \sim \tau/k \sim 10^{-3}$.

[2)]Estimates indicate that the contribution to the extinction made by this term does not exceed 1% of the values obtained in Ref. 4 and $\tau \sim 5$–15 cm^{-1}.

[3)]The propagation of light in nematic liquid crystals was investigated for $L \sim 1$–2 mm (Refs. 4 and 5). The number of defects in these samples was sufficiently low to determine the Frank moduli to within 10–15% and to measure the extinction coefficient.[4]

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